PB86-160280

Feasibility of Using Radioactive Sulfur in Long-Range Transport Experiments

Brookhaven National Lab., Upton, NY

Prepared for

Environmental Protection Agency
Research Triangle Park, NC

Jan 86

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completi--

1. REPORT NO.	2.	3. PB86-160280
EPA-600/3-86/010		

4. TITLE AND SUBTITLE	5. REPORT DATE
FEASIBILITY OF USING RADIOACTIVE SULFUR IN LONG-RANGE TRANSPORT EXPERIMENTS	January 1986
	6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
Paul Michael	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Atmospheric Sciences Division Department of Applied Science Brookhaven National Laboratory Upton, Long Island, NY 11973	CCVN1A/06-3087 (FY-86)
	11. CONTRACT/GRANT NO. IAG DW89930067-01

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Atmospheric Sciences Research Laboratory--RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711	Final 10/83 - 10/84
	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

The radioactive isotope Sulfur-35 is an ideal tracer of sulfur emissions from anthropogenic sources because it would undergo the same chemical transformations in the atmosphere as the more common isotopes of sulfur, and further it has a very low natural background concentration. A conceptual experiment using a simple transport model shows that for continual releases over a year and requiring detection at 1000 km, a source strength of about 180 curies per week is necessary. Calculations using a wind rose Gaussian plume model indicated that the air concentrations near the release point would be below Federal minimum values set for protection of the public safety. Production of the isotope is within existing technology although material costs could run as high as one million dollars for a full years release. Sampling and detection methods would require state-of-the-art technology with an investment of about one half the material costs. The general conclusion is that such a tracer experiment is technically feasible. Public perception was not considered in this study and a major effort of public education would be necessary before such an experiment could be carried out. An analysis of the possibility of using the stable isotopes Sulfur-34 and Sulfur-36 would require the development of new very expensive and specialized technology. Thus the use of stable isotopes was not considered to be feasible at the present time.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	23
	20. SECURITY CLASS (This page)	22. PRICE
	UNCLASSIFIED	

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

EPA/600/3-86/010
January 1986

FEASIBILITY OF USING RADIOACTIVE SULFUR IN LONG-RANGE TRANSPORT EXPERIMENTS

by

Paul Michael
Atmospheric Sciences Division
Department of Applied Science
Brookhaven National Laboratory
Upton, New York 11973

Interagency Agreement DW89930067-01

Project Officer

Francis Binkowski
Atmospheric Science Research Laboratory
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

ATMOSPHERIC SCIENCES RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NORTH CAROLINA 27711

NOTICE

Although the information in this document has been funded by the United States Environmental Protection Agency under Interagency Agreement DW89930067-01 to Brookhaven National Laboratory via the United States Department of Energy, it has not as yet been approved by the US EPA's required peer and administrative review and, therefore, does not necessarily reflect the view of the Agency and no official endorsement should be inferred.

ABSTRACT

Because radioactive ^{35}S has a very low natural occurrence and would undergo the same transformations as the S emitted from anthropogenic sources, it would be an ideal tracer for use in long-range transport experiments. An examination has been made to determine the feasibility of using this substance in long-range transport experiments.

A conceptual experiment consisting of continual releases of ^{35}S over a year's time period was specified in order to have as stringent as possible conditions. A simple transport model was used to estimate the release rate that would be required for detection at 1000 km would be approximately 180 curies per week. The results of a wind rose Gaussian plume model indicated that the air concentrations near the release point would be below the levels that have been set by Federal Regulations to protect public safety. Production of the radioactive material by the exposure of ^{35}Cl to neutrons in a nuclear reactor is within existing capability; the costs of the material for the extreme conceptual experiment would be substantial (of the order of one million dollars) but considered not to be so great as to be disqualifying. The sampling and detection techniques would require the use of state-of-the-art technology with an investment of about half that of the tracer material being required. The general conclusion was that the use of ^{35}S is technically feasible. However, the study did not include consideration of the public perception of the use of radioactive material on this scale. It is likely that a reasonable effort would be required for public education before approval for such experiments could be obtained.

The use of stable sulfur isotopes, ^{34}S and ^{36}S, was considered briefly. The natural occurrence of these isotopes is so large, and so variable, that very large release rates would be required (of the order of 3 tonnes of ^{34}S per hour); it was concluded that the use of stable isotopes would require the development of new technology at very large expense; until new production techniques were proven, the use of these isotopes as a tracer was not considered to be feasible.

This report was submitted in fulfillment of Interagency Agreement DW89930067-01, Task IV, from the United States Environmental Protection Agency to Brookhaven National Laboratory via the United States Department of Energy.

CONTENTS

Abstract iii

I. Introduction. 1
II. Conceptual Experimental Design. 2
III. Required Release Rate 3
IV. Allowable Release Rate. 5
V. Production. 8
VI. Detection 10
VII. Use of Stable Isotopes. 12
VIII. Conclusions 14
IX. Acknowledgments 15
X. References. 16

Preceding page blank

I. INTRODUCTION

The primary objective of the atmospheric processes research of National Acid Precipitation Assessment Program (NAPAP) is to determine how much of current acidic deposition in any part of the country is attributable to anthropogenic sources in any other regions of the country and to determine how this deposition would change in response to emission changes. This report will discuss the use of sulfur isotopes as tracers to address the first part of this objective. The fact that the primary region of interest, the Northeastern United States, has substantial concentrations of sulfur compounds both in the atmosphere and in precipitation makes it most difficult for a purely observational program to be able to unravel the source attribution problem. The approach being taken by the major fraction of the NAPAP program is to have observational and experimental programs coupled to research on the mechanisms of transport, transformation, removal, scavenging, etc., in order to be able to develop scientifically based source receptor relationships. Major field experiments are expected to provide information on processes and to give direct information on "current" source receptor relationships. It is expected that models will be the prime tool for "future" source receptor relationship determinations, and that parameterizations and verification data will come from field experiments.

A key tool for field experiments is the use of tracers as, for example, the conservative tracers employed in the CAPTEX experiment. Crucial to the source-receptor relationship is, of course, the removal processes by dry deposition, clear air chemical transformations, transformations in cloud water and precipitation, and the wet scavenging processes. These depend upon the chemical nature of the material being transported; hence, any

results obtained by the use of tracers that do not exactly mimic the pollutant of interest, in this case sulfur oxides, will, at best, require the intervention of a theoretically-based source receptor model and, at worst, be highly ambiguous. It would seem that the ideal experiment would be to attempt to either "spike" the emissions from particular source regions with stable isotopes of sulfur beyond their normal abundance, (e.g., add significant amounts of the stable isotopes ^{34}S or ^{36}S) or to release radioactive ^{35}S. This paper focusses upon the use of ^{35}S; the use of the stable isotopes is discussed briefly.

The use of ^{35}S for acid deposition studies has been suggested by others, most notably by the JASON group (Chamberlain et al., 1981) and is being considered by the groups performing feasibility studies for EPRI (MATEX) and for the US EPA. Preliminary results of this study were presented at the NAPAP public review meeting in Boston in August 1983.

^{35}S is the most suitable of the radioactive sulfur species because of its half-life, 87 days. The decay of the shorter half-lived species would be so great that a sufficient fraction would not survive the time period between production, release, transport, and measurement.

II. CONCEPTUAL EXPERIMENTAL DESIGN

The feasibility of the use of ^{35}S, or any other tracer for that matter, will depend upon the type of experiment being considered. It is beyond the scope of this report to develop a full experimental plan; instead, a broad scoping of a conceptual experiment is put forth in order to bound the feasibility. Therefore, the experiment to be used as a design basis is one that puts the tightest constraints upon the use of the tracer, e.g. requires the

maximum amount of material to be released and thus would have the maximum potential for a public health hazard.

The conceptual experiment considered, thus, consists of near-continuous releases of the tracer material from a tall stack over a year's time period with 50 or so precipitation and/or air samplers being deployed at distances the order of 1000 km from the point of release. During each precipitation event, the order of three precipitation samples would be taken; air sampling would use three-hour time intervals. The time duration of at least one year is required in order that the meteorological conditions that occur in all seasons be sampled. That the release be continual is to remove any possible bias that might result from the subjective selection of "suitable" conditions. An alternative would be releases at randomly selected times, so as not to introduce a bias in the selection of condition. However, the assumption of continual release places the most stringent conditions upon the safety calculations reported in Section IV below.

III. REQUIRED RELEASE RATE

The amount of tracer in the sample must be sufficiently larger than that which occurs naturally; in this case, radioactive ^{35}S, the natural background results from cosmic ray protons inducing a spallation reaction upon collision with Argon-40. Concentrations in precipitation have been reported by various investigators (Goel et al., 1957; Lal et al., 1960). The mean concentration is 3×10^{-10} curies (ci) per cubic meter of precipitation with a variability of about 75%; therefore, the target for the amount of tracer in a precipitation sample is to be an order of magnitude greater than the mean; that is, the target concentration in precipitation will be 3×10^{-9} ci/cubic meter.

There is data on airborne concentrations by Hicks (1969) and an extensive set by Reiter et al. (1975, 1977, 1978). The average concentrations of ^{35}S are about 3×10^{-15} ci/cubic meter of air with a standard deviation of about 60%; the target concentration for airborne concentrations should thus be ten times greater than the mean, i.e., 3×10^{-14} ci/cubic meter.

Fundamental to the calculation, or estimation, of how much tracer must be released in the course of an experiment to achieve the target concentrations is the calculation (or, again, the estimation) of the concentrations in either air or precipitation that result from a unit emission rate. There is just a bit of circularity in this; if this relative diffusion were known it would not be necessary to perform the tracer experiment. One, therefore, should use simple means of estimation and recognize that the result could be quite far from reality.

The simplest dispersion formula for air concentrations is:

$$\chi/Q = F/(U\ H \times \tan A)$$

where:

χ is the concentration,

Q is the emission rate,

U is the mean wind speed,

H is the height through which the material is mixed (assumed to be uniform),

x is the downwind distance,

A is the angle over which the material is assumed to be mixed (again, assumed to be uniform),

and F is the fraction of material not removed enroute.

Reasonable estimates are: U = 10 m/sec, H = 1000 m, x = 1000 km, and A = 10 degrees. If one assumes that the removal is due to dry deposition with a uniform deposition velocity of 1 cm/sec, the foregoing values of parameters would yield an F = 1/e = .368 and the resulting $\chi/Q = 2 \times 10^{-10}$ sec/m^3.

To calculate the concentration in precipitation, one needs a scavenging ratio of some type; for our purposes, we can assume that half the material contained in a column 1-km high is concentrated in one cm of precipitation; hence, if C is the concentration in precipitation, then the C/Q is 1×10^{-5}. (It is to be noted that this scavenging ratio of 0.5×10^5 is a factor of two less than the naturally occurring ratio of precipitation to airborne concentrations (10^5), thus, the estimated required release rate for detection in precipitation will be greater than for airborne detection by a factor of two, this larger estimate being the release rate assumed to be required.) The results of this section are summarized in Table 1.

Table 1. Required Emission Rates

		Airborne	Precipitation
Background	ci/m^3	3×10^{-15}	3×10^{-10}
Target	ci/m^3	3×10^{-14}	3×10^{-9}
Emission rate	ci/sec	1.5×10^{-4}	3×10^{-4}
Emission rate	ci/wk	90	180

IV. ALLOWABLE RELEASE RATE

The primary technical impediment to the use of a radioative tracer is the possibility that exposure to the material would cause harmful effects to members of the public in the vicinity of the release point.

There are several steps in estimating the maximum amount of material that could be released and still be considered "safe." These steps include the relationship between airborne concentration and dose to sensitive organs, the allowable dose, the relative dispersion, and the release schedule. In each step "conservative" assumptions have been made.

Models for the uptake to sensitive organs are based upon recommendations of the International Commission on Radiological Protection (ICPR). Two ICRP models of uptake were considered. The first, considered quite conservative, was proposed in 1959. It assumes that there is selective uptake by ^{35}S in the testes. The other more recent model (ICRP 1978) assumes uniform uptake and thus would allow higher airborne concentration. However, this model is not yet accepted; it is included here to indicate the level of conservatism in the estimates. Also considered were three levels of exposure: 500 mr/year to an individual; this is the limit established by the Federal Standards for Protection against Radiation (10CFR20) to which a person at a site boundary of a restricted facility may be exposed; one-third of the foregoing which is prescribed in 10CFR20 as the allowable "population dose;" and one-hundreth of the first limit. This latter is usually taken to be the target of As Low As Reasonably Practicable (ALARP) efforts. These uptake models yield the columns on the MPC table. Table 2 displays the concentrations that would be allowed using the two models and three exposure levels. It is the concentration of 9×10^{-5} ci/cubic meter (column 1, row 3) that is the conservative number for which to aim.

Annual average χ/Q calculations were performed using a Gaussian/Windrose model with BNL dispersion parameters and climatology. These calculations are conservative, i.e., error on the side of too high a χ/Q, primarily

because plume rise has been neglected. The maximum annual average χ/Q in the vicinity of a 100-meter stack is calculated to be 3×10^{-7} sec/cubic meter. This value, used with the concentrations displayed in the Table 2, gives allowable values of release rates that are shown in Table 3.

The conservative (column 1, row 3) value is that which shall be required to be less than release rate needed to perform the experiment. It

Table 2. The maximum permissible airborne concentrations of ^{35}S that would be dictated by two different uptake models and three allowable exposure levels.

	$\mu Ci/m^3$	
	Selective uptake ICRP-2 (1959)	Uniform uptake ICRP-30 (1978)
"Legal" limit to individual (500 mr)	9.0×10^{-3}	5.4×10^{-1}
Population dose (170 mr)	3.0×10^{-3}	1.8×10^{-1}
ALARP (5 mr)	9.0×10^{-5}	5.4×10^{-3}

Table 3. Release rates that would give the concentrations listed in Table 2 calculated via a Gaussian plume/wind rose model. The calculated relative diffusion (χ/Q) was 3×10^{-7} sec/m^{-3}.

	Ci/sec	
	Selective uptake ICRP-2 (1959)	Uniform uptake ICRP-30 (1978)
"Legal" limit (500 mr)	3.0×10^{-2}	1.8
Population dose	1.0×10^{-2}	0.6
ALARP	3×10^{-4}	1.8×10^{-2}

should be noted that the population dose could be reduced substantially by having multiple release points in a region, separated by several tens of kilometers; this would actually be desirable since one is really interested in a regional source-receptor relationship. Further releases spaced in time, say, every other day, etc., would further reduce annual concentrations; this depends upon the experimental concept.

V. PRODUCTION

The radioactive ^{35}S needed for the type of experiments being considered here could be produced by irradiating appropriate samples with neutrons within a nuclear reactor. Two reaction types have been considered by Lamb (1984): neutrons bombardment of potassium chloride yields ^{35}S via a $^{35}Cl(n,p)$ reaction; neutrons on ^{34}S yield ^{35}S by an (n,γ) reaction. The production via KCl irradiation was deemed to be the more desirable option because of higher yield and because ^{32}P that would be coproduced via the $^{32}S(n,p)$ ^{32}P reaction would be difficult to separate from the ^{35}S. Furthermore, the production of ^{35}S via the exposure of potassium chloride appears not to have contamination in the product from other radionuclides.

Lamb considered the feasibility and cost of producing the material in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL); he noted that the Idaho Falls Advanced Test Reactor would also have capabilities in the same range. Two HFIR positions were considered: one, the Inner Small Vertical Experiment Facilities (ISVXF), capable of handling up to 11 samples, each of 183 grams, and the Large Vertical Experiment Facilities (LVXF), capable of exposing 6 samples, of 1600 grams each, but at a lower neutron flux. He included in his estimates the cost of preparing

the target material, reactor irradiation and chemical processing to recover the ^{35}S. The first three lines of Table 4 summarizes the results of Lamb's study: the remaining lines apply his results to the idealized experiment being considered here. For the sake of feasibility considerations, an "experiment" is considered to be a week long release, repeated 52 weeks of the year. As indicated above this is an extreme requirement; it is trivial to scale these results to releases on a less stringent schedule. The technical result here is that different reactor positions would yield different optimum exposure times. The important result relevant to the feasibility of the use of ^{35}S is that the amount of radioisotope required is well within the technical capabilities of the HFIR. The dollar cost is substantial; however, it is probably within the range of any comprehensive tracer experiment.

Table 4. Sulfur-35 Production Data. Conceptual "experiment" with releases around the clock for a week, 52 "experiments" per year.

	HFIR LOCATION	
	LVXF	ISVXF
Exposure time, days	88	264
Ci/sample	640	1595
Cost/ci	$205	$135
Cost per "experiment"	$37,000	$24,000
Samples/"experiment"	3.5	4.3
Cost/year	$1,924,000	$1,248,000

VI. DETECTION

The sampling, separation and detection of ^{35}S uses well established technology; descriptions can be found in the papers by Reiter et al. (1975, 1977). Noted below are a few considerations particularly relevant to the type of experiment being considered here.

Precipitation sampling would be done using standard techniques, the only modifications that would be required result from the fact that reasonably large volumes of precipitation would be required, this will be discussed below. Thus, either larger than usual funnels would be required or multiple samples would have to be collected.

The amount of radioactive sulfur in a sample would be measured by counting the emitted ß particles in a large well shielded scintillation counter. The crucial consideration is that the detection of the radioactive sulfur be done with sufficient precision. Reiter (1975) has reported that achievement of an extraction efficiency of 80% and a counting efficiency of 25% is reasonable, and that one should expect a background count rate of about 10 cpm. A limiting source of uncertainty is the precision with which the counting efficiency is known; it will be assumed that modern techniques are such that 10% can be achieved. A 15% accuracy in the ^{35}S determination can be achieved by determining the background to the order of 1% (requiring a counting time of 800 minutes), having a 2 cpm count rate due to the sample, and counting for 6 hours. This would require a total activity in the rain sample of 5.6 microcuries; if the concentration were the 3×10^{-3} microcuries/cubic meter mentioned in Section III, then a total sample of 1.85 liters would be required; for 0.5 cm of rain, this would require either a sampler with a funnel radius of 69 cm or a number of smaller samplers

operating in parallel. For example, it would require an array of eight 25-cm samplers of the type used in the MAP3S network. (There are quality assurance advantages to the use of multiple samplers so that in any event it is likely that more than one sampler would be used at each site. In fact by having an array of samplers for which the collected precipitation would be combined prior to analysis is a means of approximating regional deposition values.) While this requirement is somewhat demanding, it is within reasonable capability. Airborne concentrations are such that the sampling requirements are less stringent, two standard Hi-Vol samplers used in parallel over three-hour time intervals would be adequate.

The counting time stated as a requirement would mean that duplicate facilities would be required. If it is assumed that each site has 80 events in the yearly period and that three samples are collected for each event, each station would yield 240 samples per year. Since the counting time for each sample would be six hours, provision for background counting, maintenance, etc. would mean that each setup could service the order of 1000 samples per year. Hence, one counting setup would be required for every three stations. If one assumes 50 stations then some 17 counters would be needed. The cost of each sampler is approximately $25,000; hence, an investment of $425,000 would be required. This is a substantial sum but is probably not excessive. Some adjustment could be made by having a less stringent accuracy requirement, sampling fewer storms, having fewer samples per event, etc.

VII. USE OF STABLE ISOTOPES

It has been suggested that stable isotopes of sulfur be considered in addition to ^{35}S. The prime advantage of using ^{34}S or ^{36}S is that one would not have to confront the problem of real or perceived public health hazard. The technical disadvantage is that while ^{35}S is purely cosmogenic, the stable isotope is emitted along with the other anthropogenic emissions, hence, somewhat larger releases will be required.

We assume here that in order that an experiment be effective, the amount of tracer material in the sample (either atmospheric or in precipitation) must be greater than the expected fluctuation in the amount of the isotope that comes from other sources. This can be seen from the following considerations. We assume that the measurements made are the total concentration of sulfur in the sample, χ, and the measured abundance of the stable isotope being used as a tracer, a_m. If a_{nat} is the natural abundance, the tracer concentration, χ_t, would be calculated as

$$\chi_t = (a_m - a_{nat})\chi$$

Excluding measurement errors, the error in this quantity will be $\sigma\chi$, where σ is the standard deviation of a_{nat}. For a truly useful experiment, the tracer concentration should exceed this quantity of a factor of N, which will be of the order of 3 to 10. Thus, we can write that the required tracer concentration should satisfy the following inequality:

$$\chi_t > N\,(\sigma/a_{nat})\,a_{nat}\chi$$

The relative error in abundance is about 1 to 5%. Thus, the following range results:

$$0.03 < N\,\sigma/a_{nat} < 0.5$$

and a reasonable estimate is about 0.15. If we are interested in precipitation quality, we note from the MAP3S network that 90th percentile for S concentration in precipitation is about 3 gm/cubic meter; if we are interested in air quality, we note that the concentrations of sulfate in an incident are of the order of 20 to 30 micrograms per cubic meter. Using the previous estimates for relative dispersion for air and precipitation concentrations, this indicates that the requirements on tracer releases for air quality are somewhat less stringent by a factor of two.

Using a sulfur concentration of 10 micrograms per cubic meter, we thus obtain the following result for the emission rate in grams/sec

$$Q > 7.5 \times 10^3\, a_{nat}$$

The following table gives the required release rates for ^{34}S and ^{36}S for continuous emissions and the amount of material required during a three-hour release. One can see that the amounts of stable isotope needed are quite substantial. Newman (1984) also has considered the use of stable isotopes; he notes that the only known quantity producer of ^{34}S is the Monsanto Corporation which produces about 200 gm/year at a cost of $500 per gram; thus a 10 tonne release would cost of the order of $500 million dollars--and would very likely require new facilities. It should be further noted that the three-hour experiment considered here is an order of magnitude less extensive than the one considered for ^{35}S.

Table 5. Emissions required for use of stable sulfur isotopes.

Mass number	34	36
Abundance	4.2%	0.017%
Required emissions		
Continuous	315 gm/sec	1.3 gm/sec
	190 tonnes/wk	770 kg/wk
3-hour release rate	945 gm/sec	3.8 gm/sec
Total amount in 3 hours	10 tonnes	41 kg.

The costs for the production of ^{36}S would likely be inversely proportional to its abundance, which is about 250 less than ^{34}S, so it would also require new technology.

VIII. CONCLUSIONS

The calculations presented here indicate that the use of ^{35}S as a tracer in long-range source-receptor experiments is feasible on technical grounds. Because of the low, natural background, the release rates required are within current technological capability and the rates are so low that no significant public health risk is expected. Costs are substantial, but are comparable with the costs of other scientific experiments. What has not been addressed here is the public perception of the use of radioactivity on the spatial scale contemplated. Consideration of the public perception issue is beyond the scope of this study; however, it is clear that time and resources would likely be required for public education.

The estimated amounts of stable isotopes required indicate that they would not be viable on the 1000-km scale.

IX. ACKNOWLEDGMENTS

Jack Shreffler and Francis S. Binkowski of EPA's Atmospheric Science Research Laboratory have served as Project Officers for this study; their support has been greatly appreciated.

I am grateful to a number of people who provided me with technical information. They include Joseph Shonka and Andrew Hull of BNL's Health Physics staff who have provided important information on the public health risk of using radioactive material; Eugene Lamb of ORNL who provided information on the production of ^{35}S; Mary Kinsely of BNL's Environmental Chemistry Division who provided information on radiochemical techniques; Bruce Hicks of NOAA's ATDL and Leonard Newman and Stephen Schwartz of BNL's Environmental Chemistry Division who provided me with information on calculations similar to those reported here.

Although the information in this document has been funded by the United States Environmental Protection Agency under Interagency Agreement DW89930067-01 to Brookhaven National Laboratory via the Department of Energy, it has not as yet been approved by the Agency's required peer and administrative review and therefore does not necessarily reflect the views of the Agency and no official endorsement should be inferred.

X. REFERENCES

Chamberlin, J., H. Foley, D. Hammer, G. MacDonald, O. Rothaus, M. Ruderman, 1981: The Physics and Chemistry of Acid Precipitation, JASON Technical Report JSR-81-25, SRI International, Arlington, VA., November 1981.

Goel, P.S., N. Narasappaya, Prabhakara, T. Rama, and P.K. Zutshi, 1959; Study of cosmic ray produced short-lived P^{32}, P^{33}, Be^{7}. amd S^{35} in tropical latitudes, Tellus XI, 91-100.

Hicks, B.B., 1969: Sulphur-35 and Beryllium-7 in ground level air at Aspendale, Nature, 224, 172.

ICRP, 1959: Report of ICRP Committee II on Permissible Dose for Internal Radiation (1959), with Bibliography for Biological, Mathematical and Physical Data. In Health Physics, Vol. 3, Pergamon Press, NY, NY.

Kinsley, M., 1985: Private Communication.

Lal, D., Rama, P.K. Zutshi, 1960: Radioisotopes P^{32}, Be^{7}, and S^{35} in the atmosphere. J. Geophys. Res. 65. No. 2, pp.

Lamb, E., ORNL, Oak Ridge, TN, personal communication, 1984.

Newman, L., 1984: An Evaluation of the Use of Sulfur Isotopes for Environmental Studies, BNL Report No. 36609.

Reiter, R., R. Sladkovic, H.-J. Kanter, W. Carnuth, and K. Potzi, 1975: Measurement of Airborne Radioactivity and Its Meteorological Application, Part IV, Annual Report, 1 April 1972 through 31 March 1973, AEC Document Number COO-3425-10, January 1975.

Reiter, R., H.-J. Kanter, R. Sladkovic and K. Potzi, 1977: Measurement of Airborne Radioactivity and Its Meteorological Application, Part VI, Annual Report 1 August 1974 - 31 July 1975, ERDA Document Number NYO-3425-14, March 1977.

Reiter, R., H.-J. Kanter, R. Sladkovic, W. Carnuth and K. Potzi, 1978: Measurement of Airborne Radioactivity and Its Meteorological Application, Part VII, Annual Report, 1 August 1975 through 31 July 1976, DOE Document Number COO 3425-16, June 1978.